



10/521839
DT12 Rec'd PCT/PTO 19 JAN 2005



INVESTOR IN PEOPLE

The Patent Office
Concept House
Cardiff Road
Newport
South Wales
NP10 8QQ

I, the undersigned, being an officer duly authorised in accordance with Section 74(1) and (4) of the Deregulation & Contracting Out Act 1994, to sign and issue certificates on behalf of the Comptroller-General, hereby certify that annexed hereto is a true copy of the documents as originally filed in connection with the patent application identified therein.

In accordance with the Patents (Companies Re-registration) Rules 1982, if a company named in this certificate and any accompanying documents has re-registered under the Companies Act 1980 with the same name as that with which it was registered immediately before re-registration save for the substitution as, or inclusion as, the last part of the name of the words "public limited company" or their equivalents in Welsh, references to the name of the company in this certificate and any accompanying documents shall be treated as references to the name with which it is so re-registered.

In accordance with the rules, the words "public limited company" may be replaced by p.l.c., plc, P.L.C. or PLC.

Re-registration under the Companies Act does not constitute a new legal entity but merely subjects the company to certain additional company law rules.

**CERTIFIED COPY OF
PRIORITY DOCUMENT**

Signed

Dated

Andrew Gessy
4 November 2004

BEST AVAILABLE COPY

THIS PAGE BLANK (USPTO)

THE PATENT OFFICE
A
19 JUL 2002
Patent Office

L02 5734682-1 D02973
1/7700 0.00-0216828.4
1/77

Request for grant of a patent

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

The Patent Office

Cardiff Road
Newport
South Wales
NP9 1RH

1. Your reference

P 100485GB

2. Patent application number

(The Patent Office will fill in this part)

0216828.4

19 JUL 2002

3. Full name, address and postcode of the or of each applicant (underline all surnames)

The BOC Group plc
Chertsey Road
Windlesham
Surrey GU20 6HJ
UK

Patents ADP number (if you know it)

7975949001

If the applicant is a corporate body, give the country/state of its incorporation

England

4. Title of the invention

Apparatus and method for fluorine production

5. Name of your agent (if you have one)

Harrison Goddard Foote

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

Orlando House
11c Compstall Road
Marple Bridge
Stockport SK6 5HH
UK

Patents ADP number (if you know it)

145 71052

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number
(if you know it)

Date of filing
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

- a) any applicant named in part 3 is not an inventor, or
 - b) there is an inventor who is not named as an applicant, or
 - c) any named applicant is a corporate body.
- See note (d))

Yes

Patents Form 1/77

9. Enter the number of sheets for any of the following items you are filing with this form. Do not count copies of the same document

Continuation sheets of this form

Description	23 /
Claim(s)	4 /
Abstract	1 /
Drawing(s)	7 + 7

CF

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (*Patents Form 7/77*)

Request for preliminary examination and search (*Patents Form 9/77*)

1 /

Request for substantive examination (*Patents Form 10/77*)

Any other documents
(*please specify*)

Covering letter with fee sheet and cheque /

11. I/We request the grant of a patent on the basis of this application.

Signature

Date

Harin Goddard Foster 18 July 2002

12. Name and daytime telephone number of person to contact in the United Kingdom

David Goddard

0161 427 7005

Warning

After an application for a patent has been filed, the Comptroller of the Patent Office will consider whether publication or communication of the invention should be prohibited or restricted under Section 22 of the Patents Act 1977. You will be informed if it is necessary to prohibit or restrict your invention in this way. Furthermore, if you live in the United Kingdom, Section 23 of the Patents Act 1977 stops you from applying for a patent abroad without first getting written permission from the Patent Office unless an application has been filed at least 6 weeks beforehand in the United Kingdom for a patent for the same invention and either no direction prohibiting publication or communication has been given, or any such direction has been revoked.

Notes

- If you need help to fill in this form or you have any questions, please contact the Patent Office on 0645 500505.*
- Write your answers in capital letters using black ink or you may type them.*
- If there is not enough space for all the relevant details on any part of this form, please continue on a separate sheet of paper and write "see continuation sheet" in the relevant part(s). Any continuation sheet should be attached to this form.*
- If you have answered 'Yes' Patents Form 7/77 will need to be filed.*
- Once you have filled in the form you must remember to sign and date it.*
- For details of the fee and ways to pay please contact the Patent Office.*

P100485GB

APPARATUS AND METHOD FOR FLUORINE PRODUCTION

5 The present invention relates to an apparatus and a method of using the apparatus for the production of fluorine.

10 Semiconductor devices, for example, are generally produced in vacuum process chambers by the chemical vapour deposition (CVD) of a plurality of layers of silicon for example. The constituent layers of material are also etched to provide a desired pattern on the device during its manufacture. Such etching in a vacuum
15 chamber results in some of the substrate material being etched, e.g. silicon, silicon oxide, silicon nitride, for example, being deposited on the process chamber surfaces over time. Most of the unused chemical reagents and by-products of the deposition or etching process are
20 exhausted from the chamber at each process step, however, some of these essentially unwanted reagents and by-products are inevitably deposited on the process chamber walls and surfaces and become potential contaminants. For example, it is possible for some of these deposited
25 materials to fall from the chamber walls and become incorporated in the devices themselves which would render them as scrap. Such unwanted deposits and residues must be periodically cleaned from the process chamber surfaces before building up to undesirable and potentially harmful
30 levels.

Although semiconductor devices have been specifically referred to above, CVD is a very widely employed technique for the production of many types of electronic

devices. For example, CVD is used in the production of thin film transistors (TFT) flat panel displays by depositing films of material on large substrates such as glass panels, for example, in the production of liquid
5 crystal displays (LCD).

Conventionally, the cleaning of undesired materials from the surfaces of CVD process chambers was accomplished by use of cleaning gases such as nitrogen trifluoride,
10 hexafluoroethane and sulphur hexafluoride. Whilst these gases work well in removing contaminants from process chambers they have the disadvantage of contributing to global warming if released into the earth's atmosphere. These gases, in use, are decomposed by plasma means to
15 release atomic fluorine in the process chamber and which is the active cleaning constituent.

More recently it has been discovered that instead of the conventional gas compounds referred to above, molecular
20 fluorine gas may be used either directly or as atomic fluorine, after treating molecular fluorine in a plasma chamber, for CVD process chamber cleaning. Molecular fluorine has the added advantage that it does not contribute to global warming. EP-A-1 138 802 describes
25 the use of molecular fluorine for the removal of contaminant materials from CVD chambers.

However, whilst EP-A-1 138 802 discussed above makes it clear that molecular fluorine in itself or treated to
30 produce atomic fluorine is efficacious in cleaning CVD chambers, there is no hint as to how such gaseous fluorine may be generated to supply a commercial CVD plant.

The electronic devices manufacturing industry, however, is not well equipped to either maintain or deal with the conventional chemical plant installations which would be required to generate fluorine in the quantities necessary for the cleaning of the number of CVD chambers which exist in a typical modern plant for the manufacture of electronic devices. The industry demands reliable, easily expandable on-site production and delivery of a high-purity fluorine gas stream. It is desirable that maintenance and expansion of a fluorine generating plant be carried out quickly and simply with minimal or no chemical hazard and importantly, no loss of production during such maintenance or expansion. The aspect of expansion is important as some users will require that fluorine generating capacity be increased as the required manufacturing capacity of electronic devices increases due to increased numbers of CVD chambers.

The use of compressed fluorine in cylinders is neither practical nor safe as there is no liquid phase with compressed fluorine even when cooled. Thus, compressed fluorine in the quantities required at a normal commercial plant represent an unacceptable environmental and safety hazard as the reactivity of fluorine increases greatly when compressed. Furthermore, the cost of providing fluorine in such a manner would be prohibitive.

On-site generation of fluorine currently exists in some industries, however, the fluorine generating apparatus has been designed to suit chemical industry standards relating to fluorine generation and requires significant on-site maintenance and frequent operator intervention and often involves handling or sampling of chemicals such as the electrolyte from which the fluorine gas is produced. This procedure is common in the chemical

industry where the expectation is that personnel will wear appropriate protective clothing and use breathing apparatus, for example, to protect themselves from hazardous fluorine gas liberated to atmosphere when cell connections are broken or cells opened. However, this approach is not acceptable in the electronics industry where the expectation is that personnel will not need to rely on protective clothing and the like and that any hazardous gases will be contained at all times and not liberated to atmosphere under any circumstances.

The use of fluorine to clean CVD tools really requires on-site generation of fluorine since the alternatives are neither safe nor economic. In the absence of a particular process benefit resulting from the use of molecular fluorine for CVD tool cleaning it has been expressed that a cost saving of at least 30% would be required for the industry to change from the existing methods and cleaning chemicals. Thus, it was originally considered that each CVD process tool might be coupled to a conventional fluorine generating cell. This approach has obvious apparent benefits in that the output of the fluorine cell may be matched to the fluorine requirements of the particular tool which it supplies. Such an approach would also minimise fluorine process piping. A further apparent advantage is that failure of one fluorine generator would not result in a plant-wide shutdown, only a shutdown of one CVD tool. However, in reality, the superficial attractions of one fluorine generator per CVD tool are outweighed by the practical and economic disadvantages of such an approach. Each fluorine generator would have to have the same process modules and would thus necessitate unnecessary and costly duplication of components and services, including: anhydrous hydrogen fluoride supply, downstream purification of the fluorine gas, gas

compression and storage and generator effluent abatement. Thus, this proliferation would result in the distribution of a relatively hazardous chemical process, including large hydrogen fluoride inventories and fluorine gas storage across an entire production plant. As a consequence of this, all fluorine generator operation and maintenance activities would be similarly distributed which would have adverse implications for plant safety.

10 Additionally, fluorine gas quality is of paramount importance in CVD cleaning applications and gas quality control is much more difficult as continuous on-line analysis is prohibitively expensive when several installations would be required and periodic fluorine gas sampling from a plurality of installations has many practical and safety drawbacks.

In contrast to the one fluorine generator per tool approach described above, the single, large conventional generator with separate feeds to a plurality of CVD tools has the overwhelming disadvantage that failure or shutdown of the generator for repair or maintenance results in the entire production process being halted for however long the generator is out of commission.

25 A first aspect of the present invention relates to apparatus for the generation of fluorine by the electrolysis of hydrogen fluoride, the apparatus comprising: a plurality of individual fluorine generating units; said individual fluorine generating unit being operably connected to a fluorine gas distribution system for the remote use and consumption of said fluorine gas; said fluorine generating units being individually isolatable from said gas distribution system and removable from the apparatus for remote maintenance.

In the above, the word "maintenance" is intended to cover any reason whatsoever for the removal of the individual fluorine generating unit from the apparatus. In the
5 present invention the word "maintenance" may, for example, include routine maintenance, servicing or repair of that cell. It is further intended that such a cell requiring maintenance be removed from the apparatus and taken to a remote site away from the plant where the
10 fluorine is being consumed such that there is no inconvenience, contamination or safety hazard to the production plant or personnel.

In this specification the apparatus is a "packaged"
15 fluorine generating apparatus. The term "packaged" is intended to mean a plant which is built and assembled at a fluorine plant supplier company, for example, tested to ensure efficient operation of the apparatus, closed down, sealed and then shipped as a self-contained plant to a
20 customer site, for example, for use of the fluorine in the customer's processing. Generally, the apparatus may be totally self contained; apart from customer provided services such as electricity, water, compressed air or nitrogen supplies, for example, to render the apparatus
25 operative; in a container shippable by land or sea and that container being the container in which the apparatus will reside and operate at the customer's site.

Packaged fluorine generating plant or apparatus may be
30 generally categorised as apparatus intended to produce in total from 0 to 2.7 kg of F_2 per hour. There exist conventional large scale fluorine generating plants in the chemical industry which are capable of producing several thousand tonnes of fluorine per year, where each
35 cell will typically generate not less than 4 kg of

fluorine per hour, these plants being able to have individual cells removed by personnel wearing and using appropriate safety equipment, they are built in-situ for a plurality of cells and associated equipment brought to site of operation and assembled. Such plants are used, for example, in the nuclear industry for the production of nuclear fuel precursors such as uranium hexafluoride, for example. Such plants are quite different and distinct from the "packaged", transportable as a unit fluorine generating apparatus forming the subject matter of the present invention. In the present invention the apparatus may be contained, for example, within a container having overall dimensions substantially not more than a standard ISO container or less as explained in greater detail below.

In this specification, the apparatus comprises a plurality of self-contained fluorine generating units. The units may be constituted by a single cell insofar as that unit has effectively one cathode and one anode. Alternatively, the unit may comprise a group of cells insofar as there may be more than one cathode and anode in that unit. Thus, the apparatus according to the present invention comprises a plurality of fluorine generating units which are isolatable one from another and from the apparatus as a whole and each unit is individually removable from the apparatus without interrupting the supply of fluorine from the apparatus as a whole. In the interests of ease of description, an individual fluorine generating unit will henceforth be termed a "fluorine cassette".

It is common terminology in the fluorine generating industry to refer to a fluorine "cell" as one metal container vessel but which vessel may possess a plurality

of anodes therein (the container itself normally constitutes the cathode). Prior art fluorine cells may typically have up to 36 separate anodes. Thus, in the present invention, each fluorine cell or cassette may have, for example, 6, 12 or 24 anodes depending upon the customer's fluorine requirements.

It is also further intended that the removed fluorine cassette be replaced with another substantially identical fluorine cassette such that the fluorine generating capacity of the apparatus is not significantly impaired.

The apparatus according to the present invention provides a self contained fluorine generation system which has sufficient fluorine generating capacity for the plant, for example as a cleaning gas for CVD chambers or tools, to which it is connected such that the total fluorine demand may be met by less than the total number of individual fluorine cassettes in the apparatus. Thus, if one fluorine cassette, for example, requires repair or maintenance or servicing then the apparatus may continue to generate the total fluorine requirement without shutdown of the fluorine generating plant or other interruption. As noted above, the removed fluorine cassette may be immediately replaced with a substantially identical (in important material and dimensional respects) fluorine cassette so that total potential fluorine generating capacity is not impaired at all. For example, for apparatus having three fluorine generating cassettes, the normal average output of the three cassettes will be less than 66% of peak output of each cassette at peak demand. Therefore, if one cassette needs to be removed for any reason, the two remaining cassettes will be able to meet the total peak fluorine demand of the application which the plant is supplying.

Thus, a single point failure of a fluorine cassette of the apparatus of the present invention does not result in shut-down of the apparatus as a whole nor a reduction in the ability to supply the peak demand of fluorine.

As noted above, fluorine generating cells have traditionally been maintained, serviced or repaired in-situ which has necessitated closing down the cell and dismantling it in-situ which, because of the extremely hazardous nature of fluorine gas and the electrolytes employed, has meant that all but essential personnel, suitably attired, must be evacuated from the area whilst the work is undertaken. Usually the down-time has been several days to complete the work. This has meant disruption and lost production time in the plant which is fed by the fluorine apparatus.

With the apparatus of the present invention, it is intended that the fluorine cassette requiring work is isolated electrically and by valve means insofar as the fluorine gas aspect is concerned, removed from the apparatus and transported, by truck for example, to a remote site where the required work is undertaken. However, a replacement fluorine cassette, held as a spare in store on-site, may be immediately installed in the apparatus. Thus, there are no time constraints on the time required for the removed cell to be brought operational again; the cell may be worked on at a site which is properly equipped for such work; no hazard is produced in respect of those people working at the plant from where the cell was removed; and, there is no lost production time at the plant.

The apparatus according to the present invention may be relatively small in terms of overall size. It may, for example, approximate the size of a standard ISO container which are used internationally for shipping and transportation of many types of goods. Such containers have an overall size of about 2.44m wide X 2.44m high X 6.5m long (or about 8' wide X 8' high X 20' long in Imperial measurements). Thus, the fluorine generating apparatus of the present invention may have a small footprint and be easily situated in a convenient location within a customer's production plant.

The apparatus of the present invention comprises a fluorine generating plant which may be shipped as a unit by land or sea. One embodiment of the present invention comprises a fluorine generating apparatus having three fluorine cassettes, fluorine purification unit, fluorine compression unit and other associated equipment all enclosed within the outer dimensions of a standard ISO container. However, there is another aspect of shipping and that is the shipping of spare or replacement fluorine cassettes; a standard ISO container may accommodate up to eight fluorine cassettes based on a width of approximately 0.74m, however, this is given as an example only as the individual fluorine cassettes may be constructed in a range of individual sizes.

The fluorine cassettes may be installed within an apparatus enclosure which may house common services for all the fluorine cassettes within that apparatus enclosure. Such common services may include fluid piping, electrical cable trunking and electrical/instrument wiring harnesses.

Hazardous fluid services to the fluorine cassettes are provided with isolation valves. Preferably, the hazardous fluid services to the cassettes are provided by double isolation valves which have a vacuum connection in
5 between. Thus, the two isolation valves may be closed and vacuum applied to remove hazardous contents before breaking the connection between the fluorine cassette and the associated piping prior to removal of the fluorine cassette from the apparatus. The vacuum extraction system
10 for removing hazardous materials is preferably connected to a scrubbing system for removing and neutralising harmful materials.

Non-hazardous fluid services may be provided by means of
15 quick-connect couplings, for example.

Once the fluorine cassette is disconnected from the apparatus it is completely sealed from the ambient environment and constitutes no danger to personnel.
20 Similarly, a new replacement fluorine cassette is also harmless until it is connected into the apparatus and the isolation valves opened to permit flow of hazardous fluids.

25 Spare fluorine cassettes may be stored on-site to enable quick replacement. Similarly, removed cassettes may be safely stored on site until they can be removed to a remote repair or servicing facility off-site, for example.

30 The fluorine cassettes may be provided with means to facilitate moving the cassette into and out of the apparatus enclosure. Suitable means may comprise wheels, for example.

35

In a preferred embodiment of the present invention the fluorine cassette may itself be provided with a self-contained enclosure around the fluorine generating cell or cells such that any leakage of fluorine gas for
5 example is contained within the enclosure. More preferably, the cassette enclosure may be connected to a vacuum extraction system which is provided with scrubbing means to remove harmful chemicals.

10 In the apparatus of the present invention each cassette has its own enclosure provided by a strong metal frame and panelling. When installed in the apparatus the enclosure is connected to extraction and scrubbing means to deal with any potential leaks, and therefore, provides
15 a sealed protective enclosure. When the cassette is removed from the apparatus, the enclosure provides the dual advantage of constituting protection for the cassette during shipping without further packaging and protection for people handling the cassette.

20 In a preferred embodiment of the apparatus according to the present invention there is also provided a fluorine purification unit through which the output of the fluorine cassettes is passed so as to remove unwanted
25 particulate material or undesirable gaseous contaminants therefrom before it reaches any process equipment for which it is destined. Such unwanted material may include hydrogen fluoride, for example, carried over from the electrolyte in the fluorine stream and which may be
30 passed through a sodium fluoride trap, for example, or carbon tetrafluoride formed by reaction of fluorine with the carbon anodes.

The fluorine purification unit may also be isolatable and
35 easily removed from the apparatus for repair and

servicing in a similar manner to the fluorine cassettes. Thus, such isolation may be accomplished preferably by double isolation valves with an intervening vacuum extraction facility as with the fluorine cassettes.

5

In a further preferred feature of the apparatus according to the present invention, there may also be provided a fluorine buffer unit connected in the fluorine line downstream of the fluorine purification unit. In effect,

10

the buffer unit collects purified fluorine being generated and holds it in tanks so as to provide a fluorine reservoir to smooth out fluctuations in supply and provide fluorine at a constant pressure.

15

The apparatus according to the present invention may all be housed within a main enclosure framework which is provided with suitable panelling so as to effectively render the enclosure sealed to the outside ambient atmosphere. Further preferably, the main enclosure is

20

provided with evacuation means so that any leakage is removed and does not contaminate the surrounding area. The evacuation system may be connected to suitable scrubbing means for the removal and safe disposal of any harmful substances.

25

The main enclosure may also preferably be provided with all of the necessary electrical power supply and electrical control systems in known manner so as to effect electrolysis of the hydrogen fluoride electrolyte to generate fluorine.

30

According to a feature of the apparatus according to the present invention the framework of the fluorine cassette may be utilised as the cathode connection of the fluorine cell or cells within the cassette thus, the mere

35

installation of the cassette within the main enclosure effects the necessary cathode connection to the electrolytic cell or cells within the cassette.

5 Preferably, the apparatus according to the present invention is further provided with purging means to remove potentially reactive fluids such as moisture, for example, from piping before fluorine is introduced. Such purging means may comprise valve means connected to
10 apparatus piping for the introduction of nitrogen, for example, into the piping so as to purge oxygen, for example, from the piping.

According to a second aspect of the present invention,
15 there is provided a method for the operation and maintenance of apparatus for producing fluorine by the electrolysis of hydrogen fluoride, the method comprising the steps of: providing a plurality of fluorine generating units operably connected to a fluorine gas
20 distribution system for the remote use and consumption of said fluorine; providing means for isolating an individual fluorine generating unit from said fluorine gas distribution system and from each other; and providing means for the disconnection of said isolated
25 fluorine generating unit from said apparatus without interruption of supply of fluorine from remaining fluorine generating units.

The same definition relating to fluorine generating units
30 is utilised in the method according to the present invention as in the apparatus according to the first aspect of the invention.

In order that the present invention may be more fully
35 understood, examples will now be described by way of

illustration only with reference to the accompanying drawings, of which:

Figure 1 shows a schematic cross sectional representation
5 of a prior art fluorine producing cell;

Figure 2A shows a front elevation (without panelling) of
a first embodiment of apparatus according to the present
invention;

10

Figure 2B shows an end elevation of the apparatus of
Figure 2A;

Figure 3 shows a side elevation of a fluorine cassette of
15 Figure 2 without enclosure panelling;

Figure 4 shows a front elevation of the fluorine cassette
of Figure 3 without enclosure panelling;

20 Figure 5 shows a side elevation of a fluorine
purification unit without panelling of the apparatus of
Figure 2;

Figure 6 shows a front elevation of the purification unit
25 of Figure 5;

Figure 7 shows a side elevation of a buffer unit without
enclosure panelling of the apparatus of Figure 2;

30 Figure 8 shows a front elevation of the buffer unit of
Figure 7; and

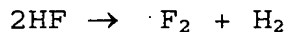
Figure 9 which shows a perspective view of a second
embodiment of apparatus according to the present

invention but with main and other enclosure panelling removed in the interests of clarity.

Referring now to Figure 1 which is intended only to explain the basic principles of the production of fluorine by electrolysis. A fluorine generating cell is shown schematically in section at 10. The cell comprises a containment vessel 12 which may or may not also constitute the cathode of the cell; in this case a separate cathode is shown at 14. The top 16 of the vessel 12 is closed save for outlets 18, 20 having valve means for fluorine and hydrogen, respectively. The vessel 12 contains an electrolyte 22 of hydrogen fluoride in a molten potassium fluoride salt. A separating skirt 24 depends from the vessel top wall 16 and its lower end 30 extends below the surface 32 of the electrolyte effectively dividing the volume above the electrolyte surface into two separate chambers 34, 36 for hydrogen and fluorine, respectively. An anode 38 generally of high-density isotropic carbon extends into the electrolyte 22 and generally extends below the lowermost extent of the skirt 24 although this may not always be the case. The vessel 12 is generally provided with means (not shown) to heat and melt the electrolyte as it is solid at room temperature. Generally, the electrolyte is maintained in the range from 80 to 100°C by the heating means when the cell is quiescent. During electrolysis heat is generated and is generally necessary to cool the electrolyte by suitable cooling means. Any suitable heating means may be used and, for example, may comprise tube heaters extending into the vessel and passing through the electrolyte, an electrically heated blanket around the vessel or a steam jacket around the vessel. A suitable power supply 40 is provided to effect

electrolysis of the electrolyte. Generally, the voltage is relatively low at about 6 to 9 volts but the current is high at about 500 to 2400 amps depending upon the number of anodes in the cassette.

5 The electrolysis reaction is:



10 The amount of fluorine generated is in direct proportion to the applied current. The gases, fluorine and hydrogen, rise substantially vertically from the anode and cathode surfaces into their respective compartments above the electrolyte surface 32. The electrolyte temperature is regulated as noted above and the composition and level is
15 controlled by the addition of anhydrous hydrogen fluoride.

Turning now to Figures 2 to 8 and where the same features are denoted by common reference numerals. Apparatus
20 according to a first embodiment of the present invention is shown at 100; the apparatus comprises a main enclosure framework 102 which has removable panelling (not shown) to form a main sealed enclosure 104 in use and which sealed enclosure is connected to a vacuum extraction
25 system (not shown) via a manifold 124 which in turn is connected to a scrubbing system (not shown) to neutralise harmful chemicals. Within the main enclosure 102, 104 are housed three fluorine cassettes 106, 108, 110 which are all substantially identical in the sense that each may be
30 substituted for another and have the same positioning of connection fittings such as isolation valves, pipes, pipe fittings, electrical services and the like, for example. The fluorine cassettes are connected to a fluorine gas manifold 114 for off-take of fluorine process gas as it

is produced by the fluorine cassettes via a fluorine gas standpipe 116 connected to the fluorine compartments of the electrolytic cells in the cassette (see Fig.1 above and also below for more detailed description of the fluorine cassette). The standpipe 116 is connected to the manifold 114 via double isolation valves 118, 120, the intervening space between which is connected to the vacuum extraction fluorine manifold 124 which in turn is connected to the scrubbing system (not shown) for neutralising any harmful gases. The hydrogen produced during electrolysis is piped away via a standpipe 130 on each cassette, the standpipes being connected via flanged joints 132 to a hydrogen gas manifold 134 which conducts the hydrogen away for either treatment or burning-off as appropriate. All of the pipe work through which fluorine flows is connected via suitable valve means (not shown) to a source of a purging gas (not shown) such as nitrogen, for example, to permit purging of oxygen and/or moisture from the pipe work prior to the introduction of fluorine.

Each individual fluorine cassette 106, 108, 110 comprises a cassette enclosure frame 140 which is able to be split horizontally into two parts: a lower part 142 which houses the fluorine generating cells 144; and, an upper part 146 which houses power supplies for electrolysis and the like. Splitting of the cassette enclosure permits easy access to the fluorine generating cells after removal of the cassette from the main enclosure 102, 104. To aid mobility, the fluorine cassette is provided with wheels 148 to facilitate removal from the main enclosure 102, 104. The cassette shown in Figs 3 and 4 has one cell producing fluorine but, however, the cell may contain 6, 12 or 24 anodes depending upon required fluorine generating capacity as explained hereinabove. The total

output of fluorine in each cassette is conducted internally to the single fluorine off-take pipe 116 having the double isolation valves 118, 120. Similarly, all hydrogen generated during electrolysis is conducted
5 to the single off-take standpipe 130. The fluorine generating cells 144 have a common containment vessel 150 fabricated from steel and forms the cell cathode and which is welded to the lower part 142 of the enclosure frame 140. Thus, the enclosure frame forms the cathode
10 connection for the whole cassette. Each cassette has its own D.C. power supply 152 and control system 154. The upper part 160 of the main enclosure 102, 104 houses busbars and main power supplies (not shown) and the like to which each of the fluorine cassettes are connected on
15 being installed into the main enclosure by means of plug-in electrical connectors (not shown) to a junction box 158.

As noted above the enclosure frame may form the cathode
20 connection of the apparatus of the present invention. Since the frame is the cathode it also carries the current which may be up to about 2400 amps with a 24 anode cassette. Thus, the frame is made of substantial section material in order to prevent undesirably high
25 temperatures being reached due to resistance heating. The cathode connection is made at 0 volts relative to earth whilst the anode connection is at 6 to 9 volts. Use of the enclosure frame as the cathode connection and current carrier enables the apparatus to be more economically
30 made with a strong frame due to thicker section materials and without unnecessary extra copper cabling to make the cathode conductors. Since the frame is a 0 volts relative to earth, the apparatus is electrically very safe.

The total fluorine output of the fluorine cassettes 106, 108, 110 is connected to a fluorine purification unit 170 through which the fluorine is passed to remove particulate material such as hydrogen fluoride or other electrolyte constituents which have been carried over by the fluorine stream and contaminants formed during electrolysis. The purification unit is shown in more detail in Figures 5 and 6. The purification unit comprises a container 172 housing chemical traps and filters (not shown) for removing unwanted material from the fluorine stream in known manner. The purification unit 170 has an enclosure framework 174 enclosing the container 172 and, in similar manner to the fluorine cassettes has a double isolation valve 178, 180 to permit installation in and removal of the purification unit from the apparatus when required. The unit is provided with wheels 180 to aid moving.

The purified fluorine gas is passed from the unit 170 to a fluorine compression unit 190 shown Figures 7 and 8. The compression unit comprises, in this example, three holding tanks 192 having a total capacity of 650 litres and able to safely withstand 5 Bar pressure of fluorine although such pressures are not generally employed with fluorine in the interests of safety. The output of purified fluorine from the purification unit 170 is fed to the compression unit pump 194 and via a pressure controller 196 to the holding tanks 192. The compression unit 190 holds a reserve of fluorine such that if for any reason the apparatus had to be closed down in respect of fluorine production for a period of time, for example, to change the purification unit 170, then there would be a reserve of fluorine to continue with the process requirements until fluorine production can be resumed. The compression unit also smoothes out fluctuations in

fluorine production so that fluorine may be supplied to the process plant at a constant pressure, for example. In similar manner to the fluorine cassette and the purification unit, the compression unit has an enclosure
5 frame 200 and wheels 202. The compression unit is connected to the fluorine manifold 114 again by double isolation valves (not shown) as with the fluorine cassette and the purification unit. The fluorine output is via a second pressure controller 198 to the fluorine
10 manifold 114 and then to the process plant where the fluorine is to be used.

As may be seen from Figures 2A, 2B, 3 and 4 the fluorine cassette 106, for example, may be installed in and
15 removed from the main enclosure 102, 104 without need to interfere with the two other cassettes 108, 110 which may continue to provide fluorine for the required processes operated externally to the apparatus. The fluorine generating capacity of the apparatus 100 is calculated
20 such that the total process requirements of the plant being served may be met, for example, by any two of the three cassettes of the apparatus described thus allowing one cassette to be redundant or removed and replaced as required.

25 In the example described above the apparatus 100 approximates less than the size of an ISO container in terms of length, and therefore, may be easily transported by land or sea. It is further possible to provide the
30 apparatus as described above but somewhat larger but still within the footprint of a standard ISO container and having empty spaces therein so as to accommodate additional fluorine cassettes for example to provide expansion of fluorine generating capacity as need arises.
35 The empty spaces may be provided with the necessary

valves and pipe connection to the manifolds etc so that an additional fluorine cassette may merely be connected into the system as with the existing cassettes.

5 The main enclosure frame 102 is provided with removable panelling so as to effect a substantially sealed enclosure, in use, to the egress of fluorine, for example. The enclosure is connected to the site extraction and scrubbing system to neutralise harmful
10 chemicals. Furthermore, each fluorine cassette, purification unit and compression unit is similarly equipped, in use, with panelling on the frames 140, 174 and 200 so as to form substantially sealed sub-enclosures within the main enclosure 102, 104, the sub-enclosures
15 also being connected to the site extraction and scrubbing system.

Figure 9 shows a single perspective view of a fluorine generating plant 300 according to a second embodiment of
20 the present invention. In terms of the capacity and capability to produce, process, control and store fluorine the apparatus of Figure 9 is similar to that described with reference to Figures 2 to 8 of the first embodiment. The apparatus 300 again has a main enclosure
25 framework 302 which is provided with panelling (not shown) to form a substantially sealed enclosure. Three fluorine cassettes 304, 306, 308 are provided, each having their own enclosure frameworks 310, 312, 314 with panelling (not shown) and each being individually
30 isolatable and removable by means of valves (not shown), as with the first embodiment. Fluorine is passed through a purification unit comprising a duty purifier 320 and a standby purifier unit 324 and then to a compression unit comprising a plurality of storage tanks 326 by means of
35 duty and standby compressors 328, 330. The fluorine is

then piped to CVD tools, for example, for use. A supply of liquid hydrogen fluoride is held in a tank 332. A hydrogen fluoride vaporiser 334 vaporises liquid hydrogen fluoride from the tank 332 and supplies it to the
5 cassettes 304, 306, 308 to maintain a constant concentration of electrolyte. A fluorine abatement module 340 is provided to remove solids from the fluorine supply, to remove fluorine from pipework when a cassette is being changed for servicing or repair, for example,
10 and extraction of fluorine not destined for customer process use. The apparatus of Figure 9 has all of the pipework purging systems, safety extraction and scrubbing systems of the first embodiment.

CLAIMS

1. Apparatus for the generation of fluorine by the electrolysis of hydrogen fluoride, the apparatus comprising: a plurality of individual fluorine generating units; said individual fluorine generating units being operably connected to a fluorine gas distribution system for the remote use and consumption of said fluorine gas; said fluorine generating units being individually isolatable from said gas distribution system and removable from the apparatus for remote maintenance, as hereinbefore defined.
2. Apparatus according to claim 1 wherein the fluorine generating units are connected to the apparatus by valve means for the isolation and disconnection of said fluorine generating units from the apparatus.
3. Apparatus according to claim 2 wherein the valve means are double isolation valves have a space therebetween which space is connected to an extraction and scrubbing system.
4. Apparatus according to any one preceding claim wherein the fluorine generating units are installed within a common apparatus main enclosure.
5. Apparatus according to any one preceding claim wherein all fluorine generating units are substantially identical to each other.
6. Apparatus according to any one preceding claim wherein said fluorine generating units are provided with wheels.
7. Apparatus according to any one preceding claim wherein each fluorine generating unit is provided with an enclosure.

8. Apparatus according to claim 4 wherein said main enclosure is connected to extraction equipment and to a scrubbing system.
- 5 9. Apparatus according to claim 7 wherein each fluorine generating unit enclosure is connected to extraction equipment and to a scrubbing system.
- 10 10. Apparatus according to any one preceding claim further including a fluorine purification unit through which the fluorine output of said fluorine generating units is passed.
- 11 11. Apparatus according to any one preceding claim further including a fluorine buffer unit connected in the fluorine line downstream of the fluorine purification unit.
- 15 12. Apparatus according to claim 11 wherein the buffer unit holds compressed fluorine.
- 20 13. Apparatus according to any one of preceding claims 7 to 12 wherein a fluorine generating cell within the fluorine generating unit is fixed to said enclosure such that said enclosure provides a cathode connection to said cell.
14. Apparatus according to claim 13 wherein said enclosure includes a framework having panelling.
- 25 15. Apparatus according to either claim 13 or 14 wherein the cathode connection is at 0 volts relative to earth.
- 30 16. Apparatus according to any one preceding claim further including purging means to remove potentially reactive fluids from piping before fluorine is introduced thereinto.
17. Apparatus according to any one of preceding claims 1 to 16 wherein the apparatus is transportable as a unit by land or sea.

18. Apparatus according to claim 17 wherein the overall size of the apparatus is at most that of a standard ISO container.
- 5 19. A method for the operation and maintenance of apparatus for producing fluorine by the electrolysis of hydrogen fluoride, the method comprising the steps of: providing a plurality of fluorine generating units operably connected to a fluorine gas distribution system for the remote use and consumption of said fluorine; providing means for isolating any individual fluorine generating unit from said fluorine gas distribution system and from each other; and providing means for the disconnection of said isolated fluorine generating unit from said apparatus without interruption of supply of fluorine from remaining fluorine generating units.
- 10 20. A method according to claim 19 wherein the plurality of fluorine generating units are provided with sufficient fluorine generating capacity such that a total demand for fluorine may be met by less than the total number of fluorine generating units within said apparatus.
- 20 21. A method according to either claim 19 or claim 20 wherein an individual fluorine generating cell may be removed from the apparatus and taken to a remote site for maintenance whilst still maintaining fluorine output to meet demand.
- 25 22. Apparatus for the generation of fluorine by the electrolysis of hydrogen fluoride substantially as hereinbefore described with reference to the accompanying description and Figures 2 to 8; or Figure 9 of the drawings.
- 30 23. A method for the operation and maintenance of apparatus for producing fluorine by the electrolysis
- 35

of hydrogen fluoride substantially as hereinbefore described with reference to the accompanying description and Figures 2 to 8; or Figure 9 of the drawings.

ABSTRACT

Apparatus and a method for the generation of fluorine by the electrolysis of hydrogen fluoride are described. The
5 apparatus comprises: a plurality of individual fluorine generating units; said individual fluorine generating units being operably connected to a fluorine gas distribution system for the remote use and consumption of said fluorine gas; said fluorine generating units being
10 individually isolatable from said gas distribution system and removable from the apparatus for remote maintenance.

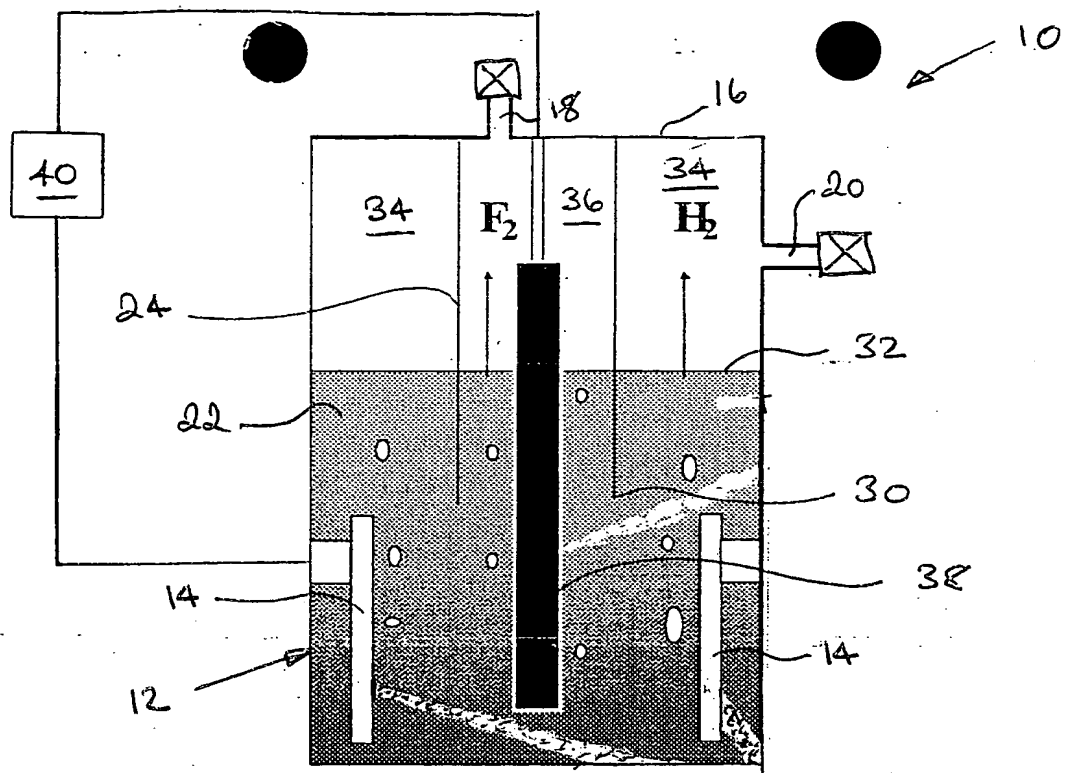


Fig. 1

THIS PAGE BLANK (USPTO)

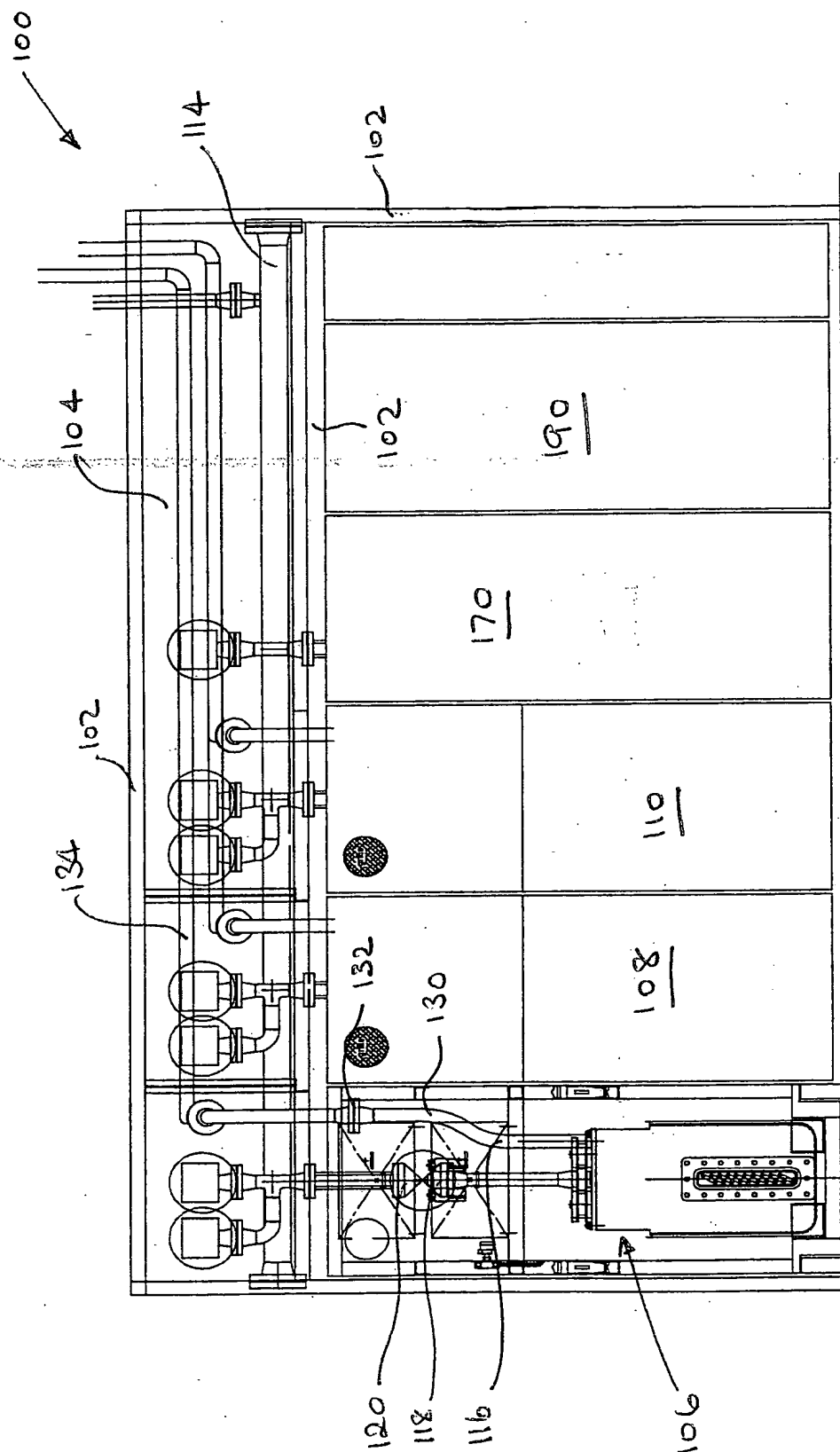


Fig. 2A

THIS PAGE BLANK (USPTO)

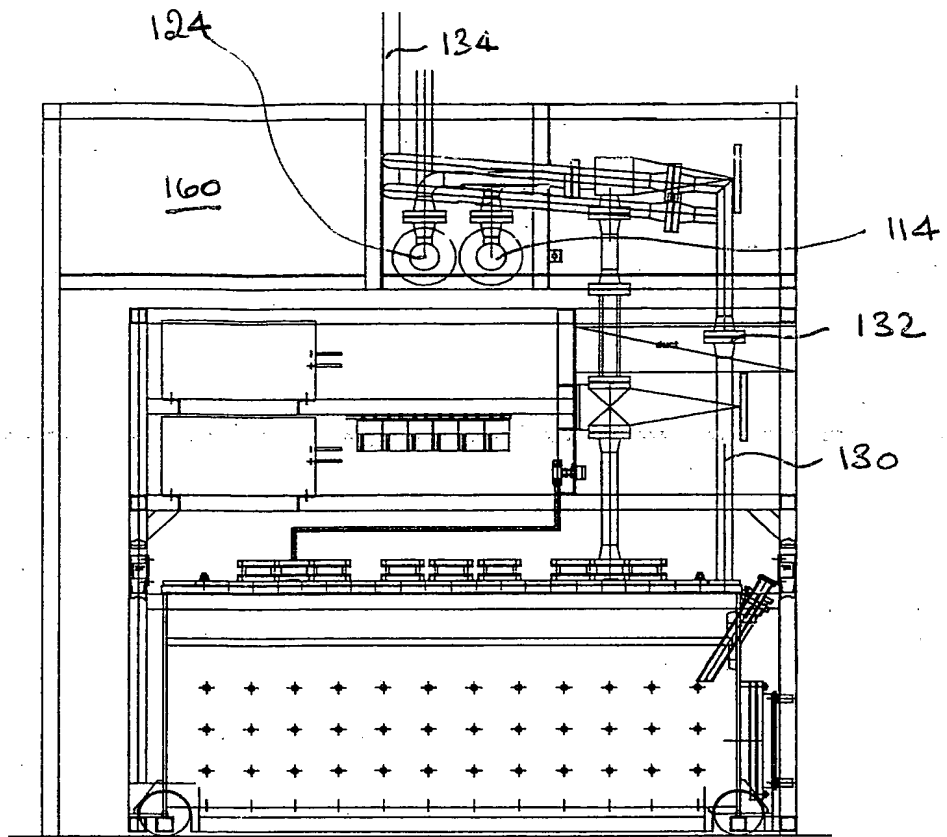


Fig. 2B

THIS PAGE BLANK (USPTO)

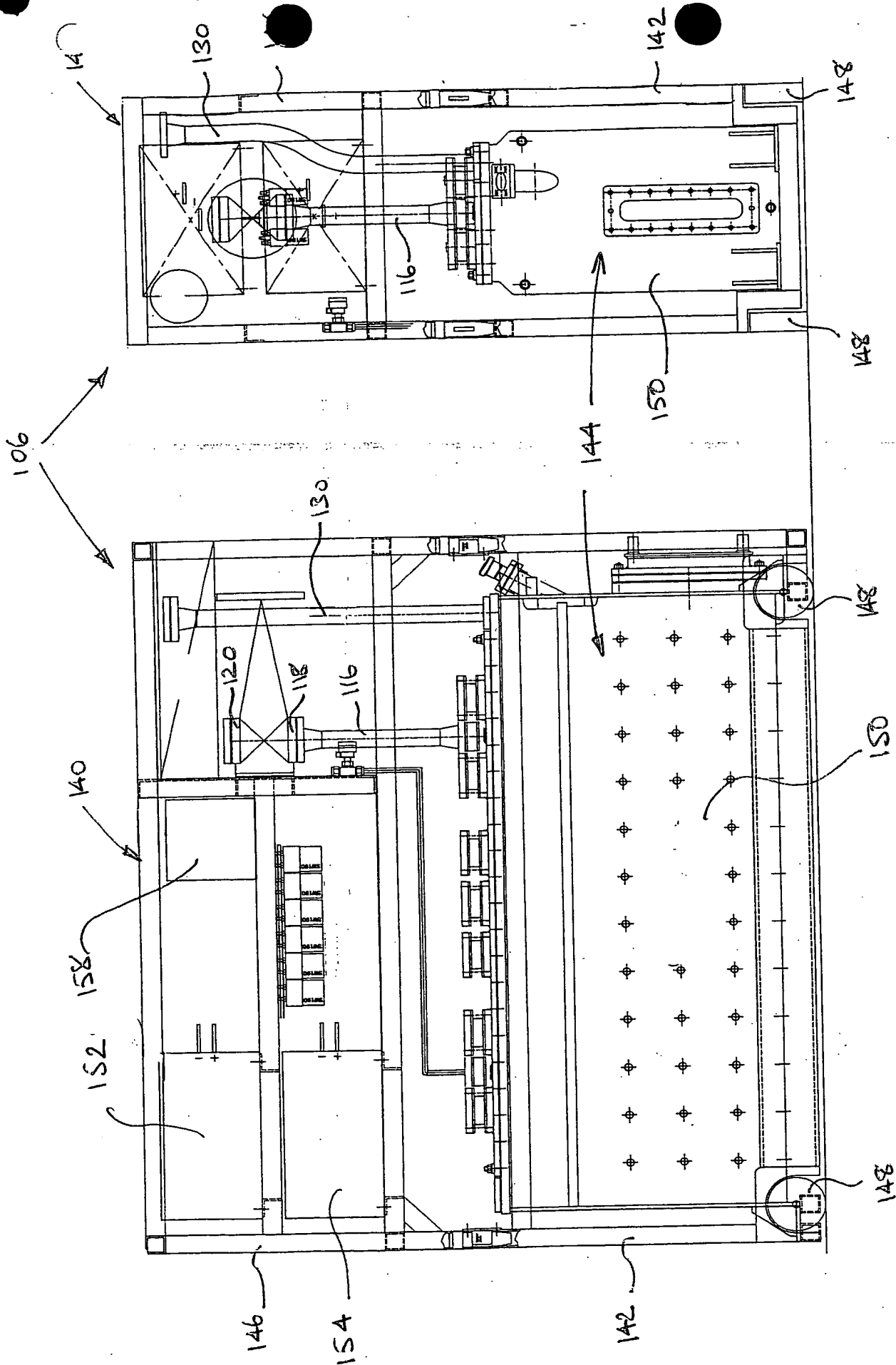


Fig. 4

Fig. 3

THIS PAGE BLANK (USPTO)

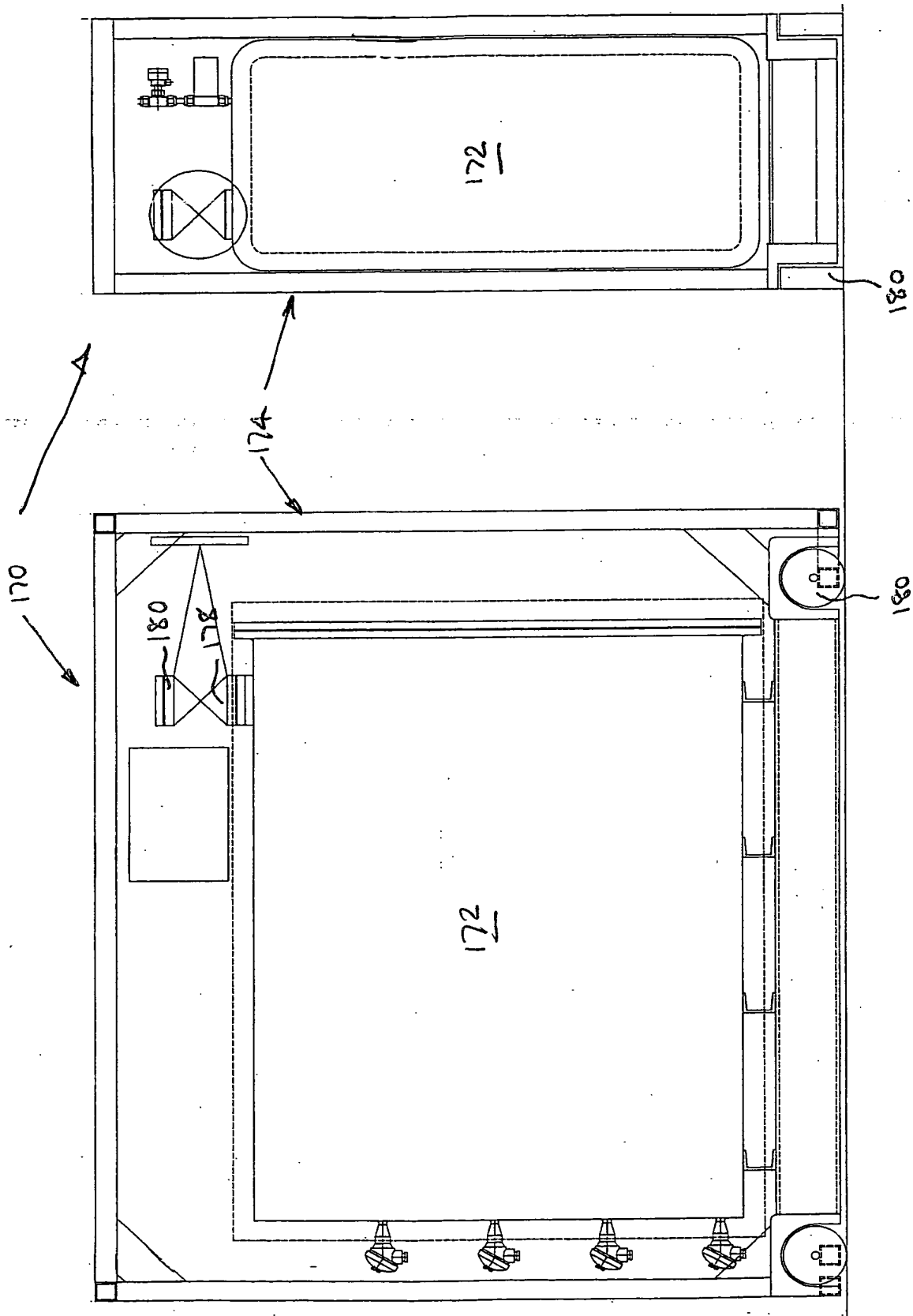


Fig. 5

Fig. 6

THIS PAGE BLANK (USPTO)

190

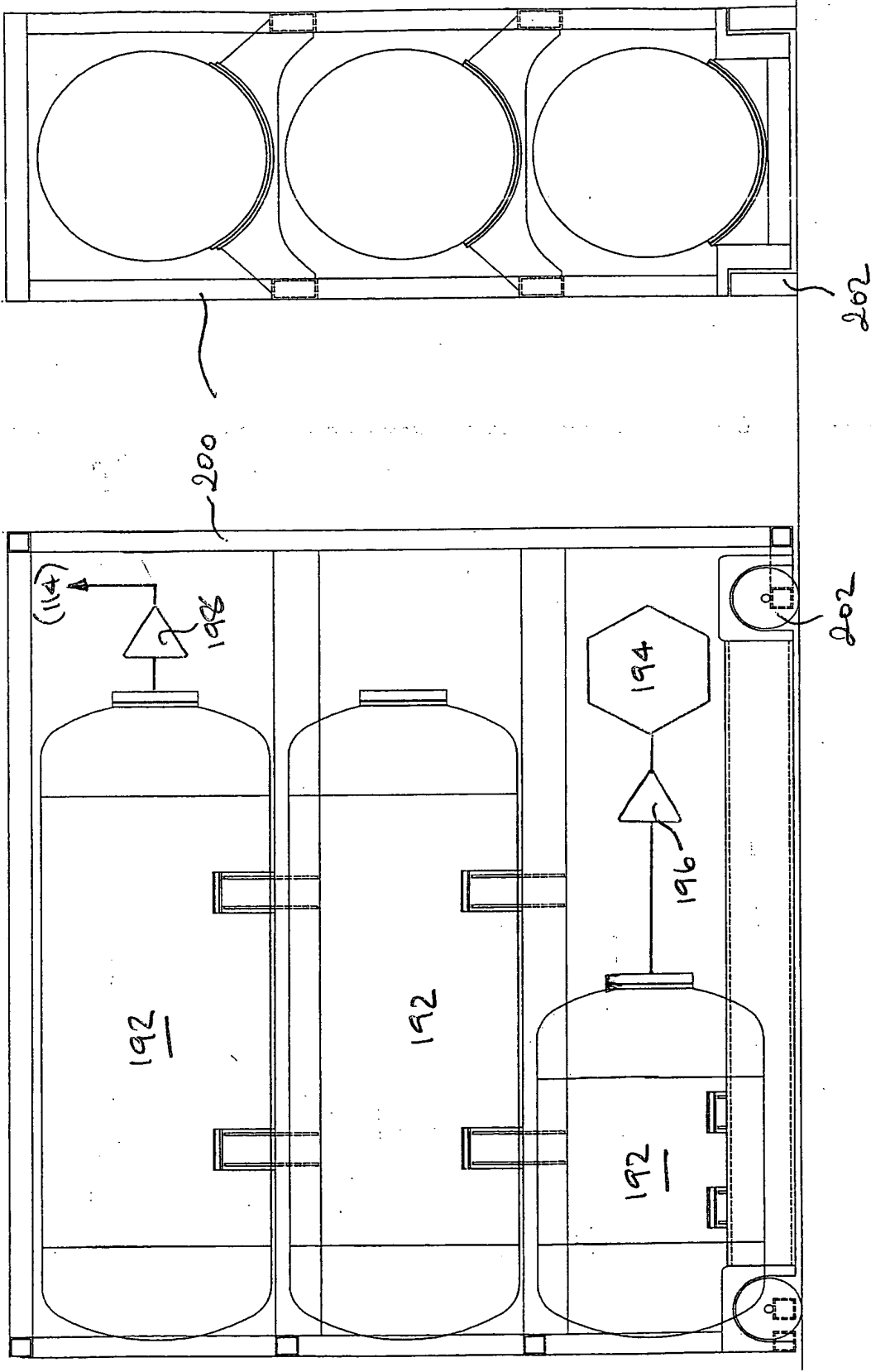
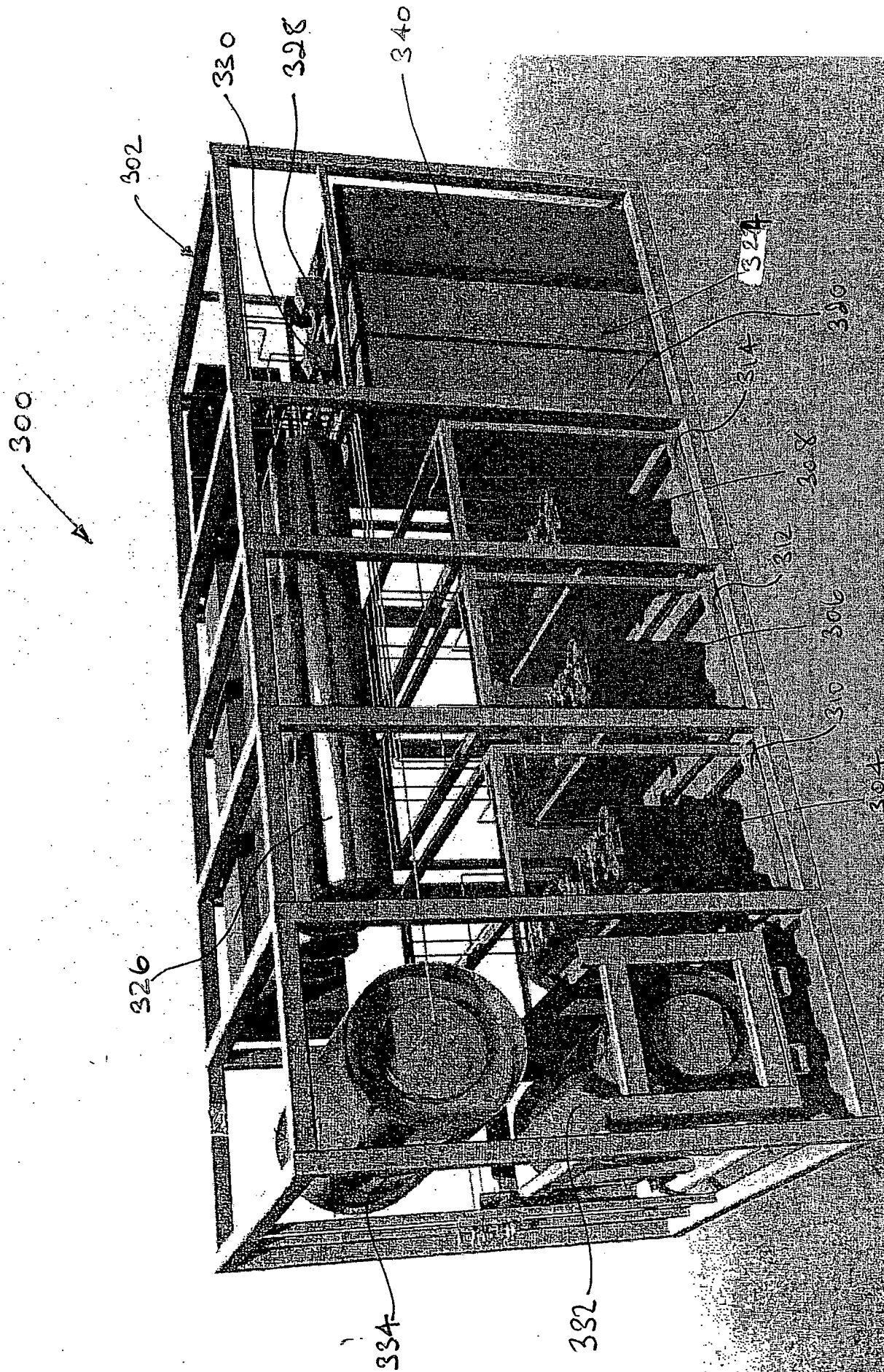


Fig. 8

Fig. 7

THIS PAGE BLANK (USPTO)



THIS PAGE BLANK (USPTO)

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☒ FADED TEXT OR DRAWING
- ☒ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☐ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.

THIS PAGE BLANK (USPTO)